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Stacking nematic elastomers for artificial muscle applications

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Abstract

Nematic liquid crystal elastomers are thermally actuated to produce macroscopic, anisotropic shape changes. Uniaxial contraction and extension of liquid crystal elastomers can be achieved by cycling the temperature of the material through the nematic to isotropic phase transition. In this report, a new approach is introduced by layering liquid crystal elastomer films to create thermally actuated stacks. A heating element and thermally conductive grease embedded between elastomer films provide a means for rapid internal heat application and distribution when a current is passed through the heating element, thus providing contractile force production in a minimal amount of time. Upon voltage application, stacks composed of two 100 μm-thick films and a single heating element produce 18% strain between contracted and relaxed states. In addition, the stacked elastomer films are capable of producing 10% contraction within 1 s and the blocked stress of the thermally actuated stacked films is calculated to be 130 kPa. The stacking approach provides new opportunities to use liquid crystal elastomers in applications requiring forces greater than those capable of being produced by single elastomer films.

Keywords: Liquid crystal elastomer; Mechanical properties; Thermal actuator; Contraction forces; Stacked actuators

1. Introduction

The development of smart materials that respond to external stimuli and result in a change in the shape or size of that material is an ever developing technology. These materials, which include hydrogels, conducting polymers, dielectric elastomers, carbon nanotube films, and nematic or ferroelectric liquid-crystalline elastomers have been developed to respond to external stimuli [1–7]. The idea of using liquid crystal elastomers (LCEs) as an actuating device, i.e. an artificial muscle, was first suggested decades ago by de Gennes [8]. In particular, uniqueness of nematic LCEs stems from the fact that they exhibit reversible macroscopic and anisotropic contraction as the material is heated and cooled through the nematic to isotropic phase transition temperature, T_{NI} [9]. The macroscopic uniaxial contraction occurs parallel to the nematic director and arises from ordered mesogenic units that are anisotropically incorporated into a polymerized and cross-linked network. The development of an optimized nematic liquid crystal elastomer depends on tight coupling and orientation of the mesogenic unit to the polymer backbone. Neutron scattering has revealed that the orientational order of the mesogenic groups is coupled to the orientational order of the polymer backbone [10,11]. The orientation of the mesogenic units with respect to the polymer backbone will partly dictate the response of the material to application of an external field. It has been demonstrated that the origin of the anisotropic, macroscopic strain response is due to the loss of side-chain mesogenic order as the material passes from the nematic to isotropic phase [7,12]. The loss of mesogenic order is accompanied by a change in the conformation of the acrylate backbone which results in macroscopic contraction of the film.

A key application issue in the use of actuators, including nematic LCEs, is the ability to produce sufficient force for particular applications given the blocked stress of the material. The solution is to either chemically manipulate the material to increase its strength or increase the cross-sectional area of the material. Chemical alteration of a nematic elastomer by increasing the cross-linking within the network will strengthen the material but the strain response is sacrificed, making an inef-

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14. ABSTRACT

Nematic liquid crystal elastomers are thermally actuated to produce macroscopic, anisotropic shape changes. Uniaxial contraction and extension of liquid crystal elastomers can be achieved by cycling the temperature of the material through the nematic to isotropic phase transition. In this report, a new approach is introduced by layering liquid crystal elastomer films to create thermally actuated stacks. A heating element and thermally conductive grease embedded between elastomer films provide a means for rapid internal heat application and distribution when a current is passed through the heating element, thus providing contractile force production in a minimal amount of time. Upon voltage application, stacks composed of two 100 m-thick films and a single heating element produce 18% strain between contracted and relaxed states. In addition, the stacked elastomer films are capable of producing 10% contraction within 1 s and the blocked stress of the thermally actuated stacked films is calculated to be 130 kPa. The stacking approach provides new opportunities to use liquid crystal elastomers in applications requiring forces greater than those capable of being produced by single elastomer films.

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fective actuator. Therefore, the other approach is to increase the cross-sectional area of the elastomer.

Previously, our group has reported the physical properties of thermally actuated liquid crystal elastomers in the form of films and fibers [6,7,13–15]. Liquid crystal elastomers are produced as thin films in order to maintain the nematic phase alignment of the LCE through the bulk thickness of the material [7]. Alignment of the nematic elastomer originates with a chemical layer at the LC interface that has been mechanically rubbed. As the thickness of a sample is increased, the alignment through the bulk material is lost, limiting the thickness of an individual LCE film. As an alternative, the cross-sectional area of a LCE can be increased by stacking films. With other materials, such as electrostrictive polymers, increased force production has been achieved by stacking or rolling sheets of material onto itself with electrodes on either side of the polymer [16]. This concept led to the idea of stacking the LCE thermal actuators, but exposed the issue of efficient heat transfer into a stack. The solution is to maximize the surface area of a stack of films in contact with a heat source, which has been demonstrated in an LCE polymer bender [17].

Actuation of a stack could be induced two ways: external heating in a temperature controlled environment or inserting heating elements in a stack to heat the LCE films internally. External heating requires that a stack be in an insulated and strictly temperature controlled environment. In addition, heating would be dependent upon the thermal conductivity of the films, i.e. the ability for heat to efficiently transfer through the individual films in order to maintain uniform contraction of a stack. Previous studies have reported electrically activated Joule heating of liquid crystal elastomers by embedding a conductive material such as graphite fibers or powder into films [18,19]. In the current study, a unique approach is described that involves internal heating of an LCE stack, whereby individual LCE films are layered between thermal grease and heating elements. The embedded heating elements in combination with a thermal grease results in rapid heat distribution through an LCE stack allowing contractile force production in a minimal amount of time.

2. Experimental

2.1. Film production

Elastomers were created with a single nematogen and two cross-linking agents as previously described [6]. The structures of these materials are schematically shown in Fig. 1. Elastomers used for stacking contained 86 mol% of the monomer, 10 mol% of cross-linker A, and 4 mol% of cross-linker B.

The procedure of preparing elastomer films has also been previously described [7]. Briefly, mixtures of monomer, cross-linker, and 0.1 mol% of the photo-initiator Irgacure-369 (Ciba Specialty Chemicals) were heated above T_{NI} and filled into a glass cell on a temperature-controlled hot stage. Glass cells were composed of rubbed poly-vinyl alcohol (PVA, Aldrich) coated glass plates, assembled anti-parallel and separated by Mylar spacers (Dupont), which determined the thickness of the

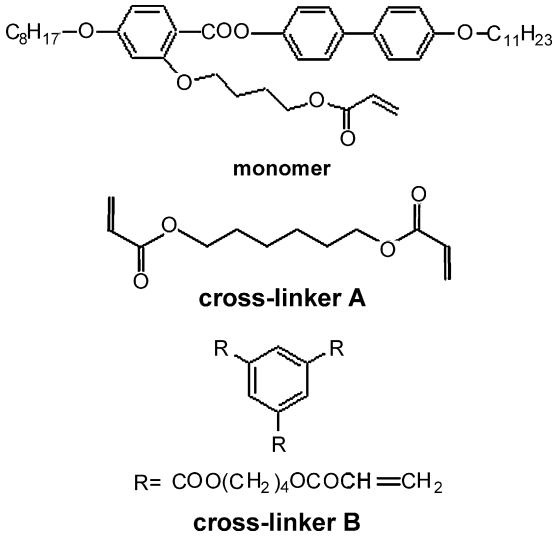

Fig. 1. Chemical structures of the components in the nematic elastomer films used for stacking, including the laterally attached nematic monomer and the two cross-linking agents.

LCE films (either 50 or 100 μm). To create LCE films, the mixture was cooled and aligned in the nematic phase and exposed to UV light for 8 min (6 mW/cm^2). Films were removed from the glass cell by dissolving the PVA in 80 °C water. Alignment of the films was confirmed by viewing the films through crossed polarizers on a microscope stage. The physical properties of individual films have been reported previously and produce uniaxial contraction of ~20% strain with a T_{NI} of 65 °C and a blocked stress of up to ~250 kPa [6].

2.2. Stack preparation

LCE films were cut into multiple pieces of the same size and coated with a thin layer of the thermally conductive compound Arctic Silver 5 (Arctic Silver, Inc.). Typical LCE film measurements were 2 cm × 1 cm ($L \times W$). The thermal grease was applied by placing a small amount near the edge of a microscope slide (to provide a uniform straight edge). The slide was drawn across the film several times to produce a smooth, even layer of thermal grease. The thickness of the thermal compound layer was typically less than 100 μm.

Heating elements were designed to fit between LCE films in order to create an internal heat source for the stacks. Ni–Cr alloy (90% nickel, 10% chromium) wire was used due to its superior thermal conductivity and common use in high temperature resistance applications. A wire diameter of 50.8 μm (0.002 in.) was used to provide the best heating while remaining embedded within the thermal grease and not significantly reducing the uniaxial strain upon actuation. A zig-zag patterning of the wire was produced by using a breadboard with evenly spaced pegs to prefabricate the heating element. This procedure provided a uniform patterned heating element to cover the maximum surface area of the LCE films. Two Ni–Cr wire heating elements are shown in Fig. 2(a). The heating elements were placed in between films to create an LCE stack, schematically shown in Fig. 2(b). Typical stack dimensions were 2.0 cm × 1.0 cm × 0.03 cm ($L \times W \times T$).

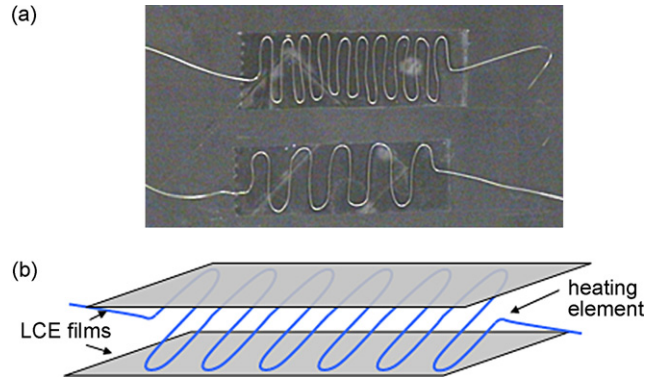

Fig. 2. Patterning of the Ni–Cr alloy heating elements and stacking of LCE films. (a) Photograph of Ni–Cr alloy wires patterned into heating elements. (b) Schematic representation of a two-film stack. Each film was previously coated with thermal grease on the side facing the heating element. In such a way, several films could be layered on top of one another.

2.3. Stack testing

Liquid crystal elastomer stacks were subjected to two types of testing to measure the strain, contraction rate, and force production. The contraction rate and strain were measured by hanging the LCE stack under minimal load (19 ± 4 mN). A ruled straight edge was placed next to the stack to monitor changes in length and experiments were video recorded for analysis. The Ni–Cr alloy wire leads were attached to a three-way switch in-line with a power supply that supplied two currents to the heating element in the stack. In order to maximize the contraction rate, a resting current was first applied to the stack to heat it to a temperature just below T_{NI} (65 °C). This current was determined by adjusting the current slightly below a level that caused contraction of the film as determined from video analysis. The resting current for stacks composed of 50 and 100 μm-thick films was determined to be 9 and 30 mA, respectively. The switch in-line with the power supply was then turned to apply a higher current and heat the stack above T_{NI} and cause uniaxial contraction. Multiple cycles of current application for each stack were recorded on video at standard frame rate and analyzed off-line. The resolution of stack length measurements used to determine the strain was 1 mm, while strain rate measurements were determined from 1-s time frames starting at the onset of the high current application.

The force produced by stacked LCEs was measured on an apparatus with a load sensor under isometric conditions [20]. The LCE stack was first mounted in a fixed clamp with the heating element attached to a power supply lead. The other end of the stack was mounted to a movable clamp with the heating element attached to the other lead of the power supply in-line with a switch. The device allowed the stack to be mounted in an isometric configuration such that the contractile force was monitored by a load cell while a current was passed through the heating elements. Stacked films were tested by systematically increasing the distance between the holding clamps following actuation (application of a current through the heating elements). Tests were performed on stacks made up of 2, 3, or 4 films stacked between thermal compound and Ni–Cr alloy heating elements. Stacks were composed of either 50 or 100 μm-thick films and the width of the stacks varied between 0.5 and 2.0 cm for all samples tested. The forces produced by the stack contraction were recorded and saved to data files via MATLAB®.

3. Results and discussion

The response of an actuator is controlled by three principle parameters: the strain of the material, the blocked stress, and the speed of the response [21]. In order to quantify the strain and contraction rate, stacks were held under minimal load and subjected to heating and cooling cycles. As shown in Fig. 3(a), the stack was suspended with two glass holders and a standard binder clip to clamp the ends of the stack. Heating was accomplished by applying a predetermined current through the heating elements, which dictated the rate of contraction. Upon current application, a rapid initial increase of the contraction of the stacks was followed by a slower, steady increase to maximal strain. As shown in Fig. 3(b), when 40 mA was applied to stacks composed of two 50 μm-thick films the contraction measured at 1 s was $4.9 \pm 0.3\%$ and $14 \pm 1\%$ at 4 s following current application. The initial contraction rate increased to $9.0 \pm 0.8\%$ within 1 s when the active current was increased to 60 mA, while the strain measured 4 s after current application remained at $14 \pm 1\%$ (Fig. 3(b)). Application of currents higher than 60 mA led to degradation and discoloration of the stack. Interestingly, the only significant difference in the contraction rate of the stacks composed of 50 μm-thick films actuated at different currents was the initial contraction observed within 1 s. This difference is primarily due to the amount of heat introduced into the system upon actuation; a higher current adds more heat to the stack system. After this initial period, the contraction at different currents was nearly identical. This response would indicate that, following the initial difference in contraction, a threshold heating limit has been reached and the stack composed of two 50 μm-thick films is absorbing heat and contracting at a maximal rate.

The contraction rate and strain of stacks composed of two 100 μm-thick films were also tested. The active current applied to the stacks ranged from 70 to 150 mA. Testing the strain over a range of currents revealed a maximal contraction of ∼18% between 100 and 120 mA. Lower currents inhibited full contraction of the films while higher currents led to film degradation. Two representative strain curves for stacks actuated under 80 and 100 mA are shown in Fig. 3(c). Stacks composed of 100 μm-thick films demonstrate 3–7% contraction within 1 s of the thermal stimulus. The strain achieved at each current consistently approached 15–20% but with differing rates, as opposed to the stacks composed of 50 μm-thick films (Fig. 3(b)). The different contraction rate for stacks with 100 μm-thick films is indicative of a limit in the rate of heat transfer through the elastomer films. While the thermal paste covers the entire surface of the film, the heat produced by the Ni–Cr alloy element must transfer across the thickness of the film itself in order to uniformly contract. Doubling the thickness of the films from 50 to 100 μm has doubled the thickness over which the heat must transfer, leading to a systematic decrease in the rate of contraction upon application of a current through the heating element.

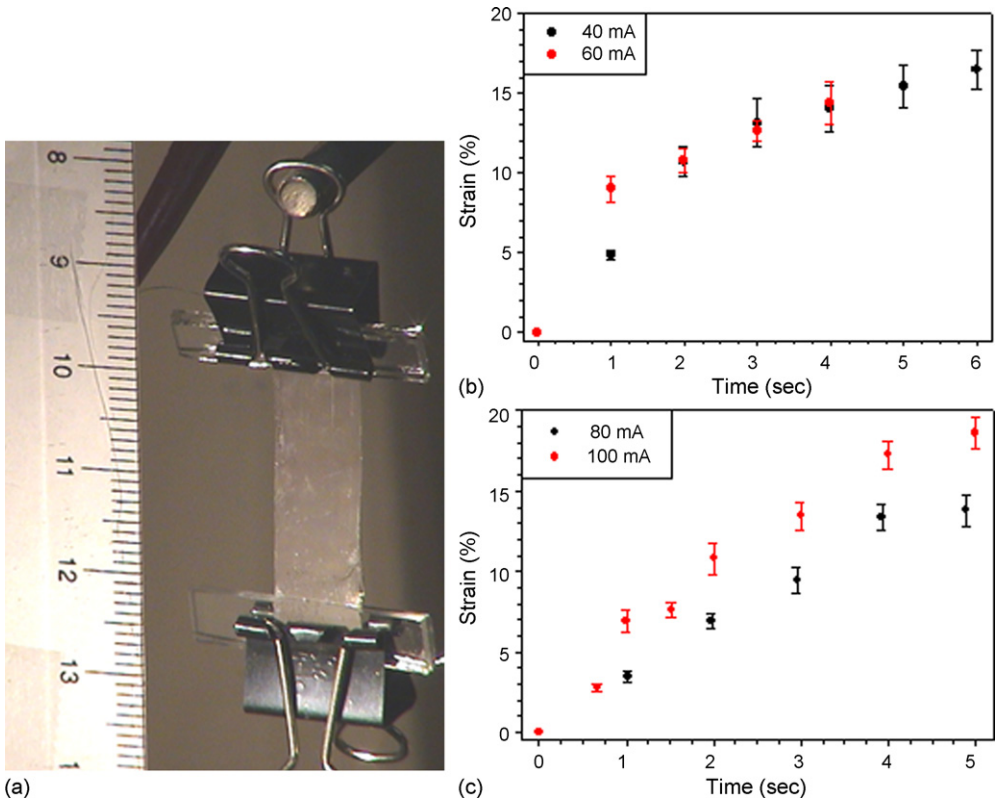

Fig. 3. Contraction rate and strain measurements. (a) Image of stack hanging under minimal load next to ruled edge. Contraction rates of stacks composed of two 50 μm (b) and two 100 μm (c) thick films. Heating currents are indicated on each graph.

The strain values observed for stacks composed of 100 μm-thick films were confirmation that the maximal strain of the LCE as a solitary film (20%) was being maintained when films were stacked with thermal grease and heating elements. Actuation of the stacks under optimized current conditions resulted in 18% strain between contracted and relaxed states over a minimum of eight cycles of 30 s heating and 30 s cooling. Within 5–10 s during these cycles the LCE stack had contracted to 80% of the full contraction. Stacks had ∼10% strain between contracted and relaxed states when a 10 s hold time was used for both heating and cooling for at least eight cycles.

The force production of stacked films was examined independent of the contraction rate on a force monitoring apparatus, as shown in Fig. 4(a). Several film stacks were tested to examine the repeatability of the force production as a function of the cross-sectional area. Under isometric conditions, force measurements represent the sustainable force exerted by a contracted LCE stack. As shown in Fig. 4(b), a stack was mounted onto holders in the force monitoring device. In order to determine the maximum sustained force of a particular stack, the distance between the holders was initially less than the length of the mounted stack (Fig. 4(b)). In such a configuration, the film was

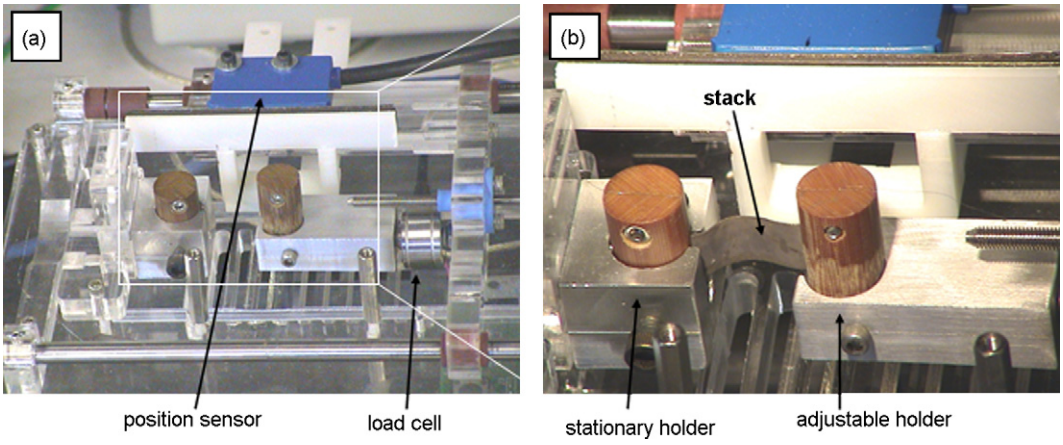

Fig. 4. (a) Image of force monitoring device. (b) Zoomed image of stack mounted on force monitoring device. As indicated, the left hold is fixed in position and the right holder can be repositioned to adjust the distance between the two holders.

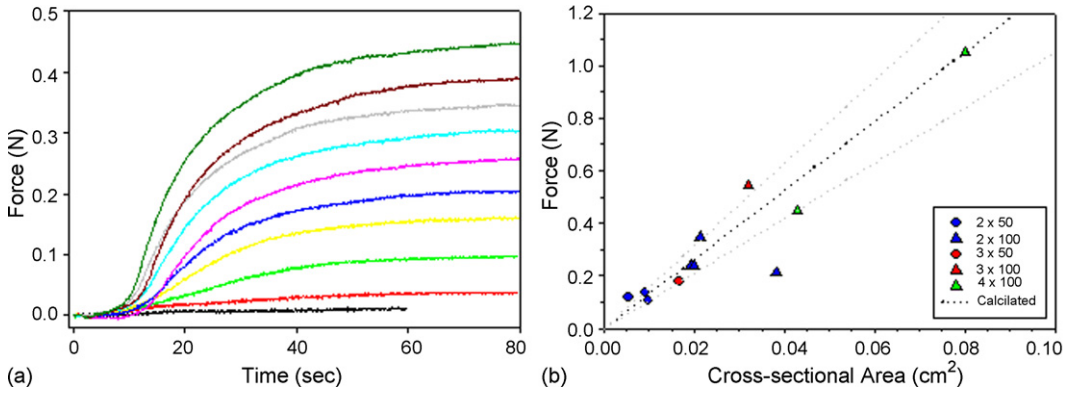

Fig. 5. Force production of LCE film stacks. (a) Maximum sustained contractile force of a four-film stack under isometric conditions. The dimensions of the stack are 20 mm × 10 mm × 0.43 mm ($L \times W \times T$). Given the cross-sectional area of the four films (4.3 mm^2), the blocked stress of this particular stack was 104 Pa. (b) Summary of the force production as a function of the cross-sectional area of the films within stacks. The key indicates the number of films (blue, red, or green) and thickness of films (circles or triangles) for each data point. The dotted lines represent the average maximum stress (black) ±20% (gray) produced by the stacks. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

initially free to contract upon application of a current. As contraction progressed, the slack was reduced until the stack began to exert a contractile force on the holders, which was monitored by a load cell aligned with the adjustable holder (Fig. 4(a)). In successive runs, the distance between the two holders was incrementally increased, reducing the slack during contraction and increasing the force exerted on the load cell. Fig. 5(a) shows the force profile of a stack composed of four 100 μm-thick films as the displacement between the stack holders was incrementally increased each run. In this stack, three heating elements connected in parallel were used to apply the thermal stimulus. The maximum sustained force prior to failure of the stack was 0.45 N. Ultimately the stack yielded when the force produced by the stacks was overcome by the force required to keep the holders stationary. Note that the equilibrium force produced by the stack increased over a time frame much longer than the times reported for experiments examining the contraction rate. In the force monitoring device, stacks were held at room temperature prior to application of a current with no resting current being passed through the heating elements.

The relationship between the cross-sectional area and the maximum force production was investigated for several stacks containing either 50 or 100 μm-thick films of varying widths. If the integrity of single films within the stacked LCEs is maintained, it is expected that the maximum force and cross-sectional area of the stacks should collapse onto a single curve. As shown in Fig. 5(b), this expectation is realized and the average blocked stress of the stacked LCEs was calculated to be 130 ± 40 kPa. The maximum force production for the stacked LCEs ranged from 0.1 N for a stack composed of two 50 μm-thick films to over 1.0 N for a stack composed of four 100 μm-thick films. Given the cross-sectional area of a particular stack, the maximum sustained force can be estimated over the 10-fold range of forces observed in the current study.

4. Conclusion

It has been demonstrated that thermally actuated LCE films can be stacked into multi-layered units for tunable force production over a 10-fold range. The incorporation of heating elements provides a unique way to internally heat a stack of LCEs to induce uni-axial contraction upon application of a current. The thermal grease serves a two-fold purpose of inhibiting film degradation in areas in contact with the heating element and distributing the heat over the surface area of the film. The method of stacking eliminates the need for an externally controlled environment for applications ranging from robotics, microfluidics, shape changing membranes, deformable fins for autonomous underwater vehicles, etc. Future studies include examining threshold contraction rates and force production as a function of the number of films in a stack and designing ways to incorporate the stacked films into mechanical devices.

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Christopher M. Spillmann is a research physicist at the Center for Bio/Molecular Science and Engineering, Naval Research Laboratory (NRL), Washington, DC. He received his BA in physics from the College of Wooster in 1997 and his PhD in biophysics from the University of Rochester in 2004. In 2004, he was awarded a National Research Council Associateship at the NRL. He has expertise in the field of cellular mechanics, receptor/ligand interactions and liquid crystal applications. His recent work with liquid crystals includes actuators and functionally active nano-materials.

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Waleed Farahat received his BS degree from the American University in Cairo, Cairo, Egypt, in 1997, and his SM degree, in 2000, from the Massachusetts Institute of Technology (MIT), Cambridge, where is currently working toward his PhD degree in mechanical engineering. His current research interests include the application of dynamical systems, control, and optimization theories to neuromuscular control, rehabilitation, and robotics.

Hugh Herr received his BA degree in physics from Millersville University, Millersville, PA, in 1990, his MS degree in mechanical engineering from MIT, Cambridge, and his PhD degree in biophysics from Harvard University, Cambridge, MA, in 1998. He is Assistant Professor of the Media Laboratory at MIT, Cambridge, and the MIT-Harvard Division of Health Sciences and Technology, Cambridge. His primary research objective is to apply principles of muscle mechanics, neural control, and human biomechanics to guide the designs of biomimetic robots, human rehabilitation devices, and augmentation technologies that amplify the endurance and strength of humans. He has advanced novel actuation strategies, including the use of animal-derived muscle to power robots in the millimeter to centimeter size scale. In the field of human rehabilitation, his group has developed gait adaptive knee prostheses for transfemoral amputees and variable impendence ankle-foot orthoses for patients suffering from drop foot, a gait pathology caused by stroke, cerebral palsy, and multiple sclerosis.

Banahalli R. Ratna is currently the Head of the Laboratory for Interfacial Interactions at the Center for Bio/Molecular Science and Engineering, NRL, Washington, DC. She received her PhD in physics from Mysore University, India. Her research interests include physics and application of liquid crystal and directed colloidal self-assembly. She has more than 140 research publications and patents. Since she joined the NRL in 1994, she has studied various electro-optic properties of liquid crystals, such as electroclinic and pyroelectric effects. Her interests also include the development of novel bio-based nanostructures. Currently, she is leading a group engaged in developing liquid crystal elastomers for artificial muscle applications and active nano-materials.